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**On the Phase Shift of Reflection High Energy Electron Diffraction Intensity
Oscillations during Ge(001) Homoepitaxy by Molecular Beam Epitaxy**

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Abstract

We have conducted a systematic investigation of the phase shift of the Reflection High Energy Electron Diffraction (RHEED) intensity oscillations during homoepitaxy of Ge(001) by molecular beam epitaxy for a wide range of diffraction conditions. Our results show that for small incidence angles with a beam azimuth several degrees away from the $\langle 110 \rangle$ crystallographic symmetry direction, the phase is independent of incidence angle; however, it starts to shift once the incidence angle is high enough that the (004) Kikuchi line appears in the RHEED pattern. Moreover, under some conditions we observe the oscillations from only the Kikuchi feature and not from the specular spot, and the oscillatory behavior of the Kikuchi feature is almost out of phase with that of the specular spot. We conclude that the phase shift is caused by the overlap of the specular spot and the Kikuchi features, in contrast to models involving dynamical scattering theory for the phase shift. We discuss necessary conditions for avoiding interference.

I. INTRODUCTION

Due to its high surface sensitivity and its compatibility with systems for UHV thin film growth by methods such as Molecular Beam Epitaxy (MBE), Reflection High Energy Electron Diffraction (RHEED) has been widely used in monitoring the surface structure and the quality of a film during epitaxial growth.¹ Particularly, the observation of the intensity oscillation of a specularly reflected spot during growth, since it was first reported in the early eighties,² is routinely used in measuring the deposition rate and determining the film growth mode. Despite its popularity, fundamental questions about the origin of the oscillation remain.³ Among the proposed models, the kinematic approximation⁴ and the phenomenological step-density model⁵ are the earliest ones and are still commonly applied to interpreting RHEED results. Both models assume that the RHEED intensity is determined by a single parameter of the evolving morphology such as layer coverage or step density. However, these models fail to explain the dependence of the RHEED intensity oscillations on diffraction conditions, presumably due to the lack of consideration of multiple scattering, which is believed to be important during RHEED. An example of diffraction conditions affecting the intensity oscillation is the phase shift, termed the $t_{3/2}/T$ phenomenon,⁶ in which the position of the 1st peak (or, equivalently, the 2nd minimum as used in Ref. 6) of the intensity oscillations changes with the incidence angle of an electron beam while the periodicity stays unchanged.^{6,7} The phase shift phenomenon has led to the use of dynamical diffraction theory accounting for multiple scattering. Such calculations are, in some cases, qualitatively consistent with the measurements,^{8,9} but significant discrepancies continue to be unexplained.^{1,10} Therefore, we have conducted a systematic investigation of the phase shift of the RHEED intensity

oscillations under a wide range of diffraction conditions during homoepitaxial MBE growth of Ge(001), our prototypical system. Our results convincingly demonstrate the importance of the Kikuchi features in influencing the RHEED oscillations, an effect that has not attracted much attention in interpreting RHEED results until now.

II. EXPERIMENT

An electron beam of 15 keV is employed for the RHEED measurements. RHEED patterns imaged on a phosphor screen are transferred by a charge coupled device (CCD) camera with a temporal resolution of 1/30 sec. into a commercially available software package¹¹ for data collection and analysis. All the data presented in this article are obtained from Ge(001) homoepitaxy by MBE at 100°C. The details on substrate preparation and MBE growth are reported elsewhere.¹² The incidence angle of the electron beam ranges from 0.5° to 4° from grazing. Azimuthally, the electron beam is directed along $\langle 110 \rangle$, 7° off, or 15° off from $\langle 110 \rangle$. Prior to every intensity oscillation measurement, a buffer layer of 20 nm is grown at 370 °C to provide a smooth starting surface. Atomic Force Microscopy reveals that the starting surface consists of terraces of an average size of ~150 nm separated by steps running along $\langle 100 \rangle$.

III. RESULTS AND DISCUSSION

A set of intensity oscillations taken at 7° off the $\langle 110 \rangle$ azimuth with various incidence angles is shown in Fig. 1(a). The incidence angle is denoted near each intensity oscillation curve. In the context of the kinematic approximation, for a Ge(001) surface the incidence angles of 0.99° and 1.98° correspond to the out-of-phase and in-phase

conditions, in which electrons reflected off terraces separated by a single atomic height step interfere destructively and constructively, respectively. For all incidence angles shown, the period of the first oscillation is shorter than that of succeeding ones. This occurs because, upon opening the shutter of the effusion cell containing MBE source materials, the temperature of the cell drops rapidly to a new steady-state value. During this transient, the deposition rate is higher and consequently the period of the first oscillation is shorter.¹³ From Fig. 1(a) it is apparent that both the amplitude and the phase of oscillations are similar for the two lowest incidence angles, but a further increase in the incidence angle results in a noticeable decrease in the amplitude and change in the phase. For a quantitative analysis of the phase shift, following Zhang et al.⁶ we define the phase of oscillations as the time of the 2nd minimum ($t_{3/2}$) divided by the steady-state period (T). In Fig. 1(b) we show the phase of the oscillations presented in Fig. 1(a) as well as measurements performed at 15° off <110> and along <110> azimuth. The results from 7° off and 15° off the <110> azimuth are nearly identical within the experimental uncertainty. Both exhibit a plateau for small incidence angles, roughly up to 1.0°, and the phase decreases remarkably for greater angles. Several models involving dynamical scattering theory have been developed in order to explain the phase shift.⁸⁻¹⁰ In the dynamical calculations, some approximations are made for the scattering potential of a material upon which an electron beam is incident. Depending on the details of the potential used, contradictory results have been obtained. For example, an increasing step density is predicted to increase the RHEED intensity,¹⁴ not to affect it significantly for a wide range of conditions,⁹ or decrease it.¹⁵ The essential feature explaining the phase shift in all of these dynamical scattering models is the proportional potential, in which the

scattering potential of a growing layer is proportional to the layer coverage and approaches the bulk inner potential at the completion of a monolayer (ML).¹ In this case, the RHEED specular intensity is determined by the interference between electrons reflected off the surface and those refracted into the growing layer and reflected from the growing layer-subsurface interface.⁸ The dependence of the potential on the layer coverage, i.e., the change in the refraction condition with the coverage, allows an intensity minimum to occur at a coverage other than half a ML. The predicted phase from the simple potential model (the simplest of the proportional potential model)¹ is included in Fig. 1(b). The general trend of decreasing phase with increasing incidence angle seen experimentally is reproduced using this model. Although a better fit could be achieved by shifting down the values from the model to compensate for the artificially smaller periodicity of the first oscillation in the experimental results due to a transient high flux upon opening the shutter, the model fails to explain the plateau at low incidence angles.

Nemcsics approached the phase shift phenomenon through the dependence of the surface coherence length of an electron beam on the incidence angle.¹⁶ The main argument is that one should start to see the phase shift when the surface coherence length becomes smaller than an average terrace length of the sample as the incidence angle increases, i.e., not all of the electrons falling on the entire terrace are coherent. In our case, the estimated surface coherence length using the formula given in Ref. 14, $\lambda / (2\beta \sqrt{1 + (\Delta E / E)^2} \sin \theta)$, where θ is incidence angle from grazing of an electron beam, λ is the wavelength of the incident electron, β and ΔE are the angular and energy spread of the beam, respectively, is four times as large as the average terrace length of the starting surface, ~150 nm, for the incidence angle where the phase shift starts to occur.¹⁷

This rules out the scenario presented by Nemcsics as an explanation of the phase shift behavior documented here.

To investigate the onset of the phase shift near 1.1° we examine the actual RHEED patterns, shown in Fig. 2. In going from Fig. 2(a) to 2(d), the incidence angle is increasing as indicated in the right bottom corner of each image. It should be noted that rather longer exposure times were used in acquiring images shown in Fig. 2. There is a detectable difference between Fig. 2(a) and 2(b). The specular spot in Fig. 2(a) is circular, showing no sign of anisotropic momentum transfer of diffracted electrons. In contrast, we observe in Fig. 2(b)-(d) a streak parallel to the shadow edge, which indicates an (00n) type Bragg reflection from the bulk. This streak is the (004) Kikuchi line from the bulk. Two observations regarding the (004) Kikuchi line in Figs. 2(b)-(d) are noteworthy. Firstly, the relative position of the (004) Kikuchi line from the shadow edge varies with the incidence angle, which was unexpected because, in principle, the distance from the shadow edge of a line from bulk diffraction should not be affected by any change of the incidence angle. Secondly, the Kikuchi line is superimposed on the specular spot for the incidence angles of 1.4° - 2.8° and it can be separated from the specular spot above $\sim 3^\circ$ as in Fig. 2(d). These observations can be understood as follows. The formation of the Kikuchi line in RHEED is from bulk Bragg reflections of electrons inelastically scattered into a bulk with a typical energy loss of a few tens of eV,¹⁸ as if there were a point source of electrons emitting over a range of directions within the bulk. Actually, however, there is a strong tendency of forward scattering,¹⁹ implying a majority of electron flux scattered inelastically into the bulk still makes a grazing angle with respect to (004) planes and, therefore, only samples a small depth below the surface. This would cause bulk

reciprocal spots to be elongated along the surface normal and, therefore, relax the (004) Bragg condition. For example, if the sampling depth is 1 nm, the deviation of the (004) Bragg angle can be as much as 0.5° . Moreover, the energy loss by the inelastic scattering accompanying the formation of the Kikuchi line further helps to relax the Bragg condition by increasing the thickness of the Ewald sphere. The position of the (004) Kikuchi line – or the exit angle of electrons leaving the sample – is determined by the product of the relaxed Bragg condition, which may be described by a sinc function, and the electron flux available, which is mostly forward scattered.

The commencement of the phase shift once the (004) Kikuchi line appears leads us to conclude that the phase shift is related to the Kikuchi features. Our conclusion is further supported by the striking drop in $t_{3/2}/T$ in Fig. 1(b) upon moving to the $\langle 110 \rangle$ azimuth, where many Kikuchi lines other than (004) interfere with the specular spot for almost any incidence angle. The confounding of the interpretation of RHEED oscillations by Kikuchi features in GaAs growth has been suggested previously by Zhang *et al.*,⁶ Crook *et al.*,²⁰ and more recently by Tok *et al.*³ However, subsequent theoretical works^{8-10,21} have rejected this interpretation and offered general explanations for the phase shifts based solely on dynamical diffraction theory with various models for the scattering potential of the growing layer. Our results are definitely inconsistent with these models -- especially the absence of the phase shift at the lower incidence angles, contrary to all dynamical calculations of which we are aware -- and support the earlier general picture in explaining the phase shift.^{6,20}

Given the influence of the Kikuchi features on the measured specular intensity, it remains to be determined how they affect the phase of the oscillations. To answer this

question, we measured how the intensity of the (004) Kikuchi feature by itself varies. Fig. 3 shows the intensity oscillations of the specular spot, A in Fig. 2(d), and the (004) Kikuchi feature, B in Fig. 2(d), recorded separately during growth. Interestingly, we observe an oscillation only from the Kikuchi feature.²² Furthermore, the oscillatory behavior of the Kikuchi feature is almost the opposite of that expected of the specular spot, i.e. an initial drop in the intensity followed by a recovery as layer coverage becomes close to the completion. This may be explained by a higher rate of electron scattering into the bulk at a rough surface, which contributes to the bulk diffraction.¹⁹ Indeed, the Kikuchi oscillation not being in phase with the specular spot oscillation has been reported previously.^{19,20} Except for the very low incidence angles, below $\sim 1.1^\circ$ for the case of Ge(001) used in this study, the RHEED intensity measurement is always affected by the presence of the Kikuchi features located very close to the specular spot. In this case the RHEED intensity variation may be considered to be the superposition of two oscillations – that of the specular spot and of the Kikuchi feature – with different phases and amplitudes. If the Kikuchi features oscillate similarly at other incidence angles to their behavior in Fig. 3, then as the amplitude of the intensity oscillation of the specular spot becomes smaller with increasing angle by moving away from the out-of-phase condition as expected from the kinematic approximation, oscillations of the Kikuchi features become an increasingly significant contribution, thereby leading to the more pronounced phase shift.

IV. CONCLUSION

We have demonstrated that for Ge(001) RHEED oscillations, the phase shift is caused by the overlap of the specular spot and the Kikuchi features. The results are inconsistent with all models to explain the phase shift based on dynamical scattering theory of which we are aware. We have shown that the most surface-sensitive specular spot can be readily affected by Kikuchi features under certain diffraction conditions and that the absence of the phase shift is a necessary condition for avoiding interference. Therefore, if one is to use the RHEED specular intensity oscillation to learn about surface morphology, one must be extremely careful that the RHEED measurements be conducted under conditions where the influence of the Kikuchi features is minimal.

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- ²² It is not understood why the specular spot intensity does not exhibit oscillations. The incidence angle used is close to the second order out-of-phase condition so, according to the kinematic approximation, the strongest oscillation is expected. Furthermore, under the same growth conditions we observe specular spot oscillations at low incidence angles.

FIGURE CAPTIONS

Fig. 1. (a) RHEED intensity oscillation taken at 7° off $\langle 110 \rangle$ azimuth with various incidence angles as indicated near each trace. Zero on time axis is when shutter is opened. Intensity is normalized by pre-deposition value. Position of second minimum in intensity, $t_{3/2}$, and steady-state period, T , are also indicated. (b) Plot of phase, $t_{3/2} / T$, vs. incidence angle for oscillations collected along $\langle 110 \rangle$, 7° off, and 15° off $\langle 110 \rangle$ azimuth. Calculation using the simple potential model, discussed in text, is also included as a dashed line. Solid line is simply a guide to the eye.

Fig. 2. RHEED patterns taken at azimuth of 7° off $\langle 110 \rangle$ for incidence angles of (a) 0.99° , (b) 1.40° , (c) 1.75° , and (d) 3.15° prior to deposition at 100°C . Spot on right of each image is straight-through beam and spot on left is specular spot. Dotted line separating these spots is shadow edge. CCD exposure time used was (a) 7/30, (b) 10/30, (c) 10/30, (d) 15/30 second.

Fig. 3. Plot of intensity evolution of specular spot (A in Fig. 2(d)) and (004) Kikuchi feature (B in Fig. 2(d)). Zero on time axis is when shutter is opened.

Fig. 1

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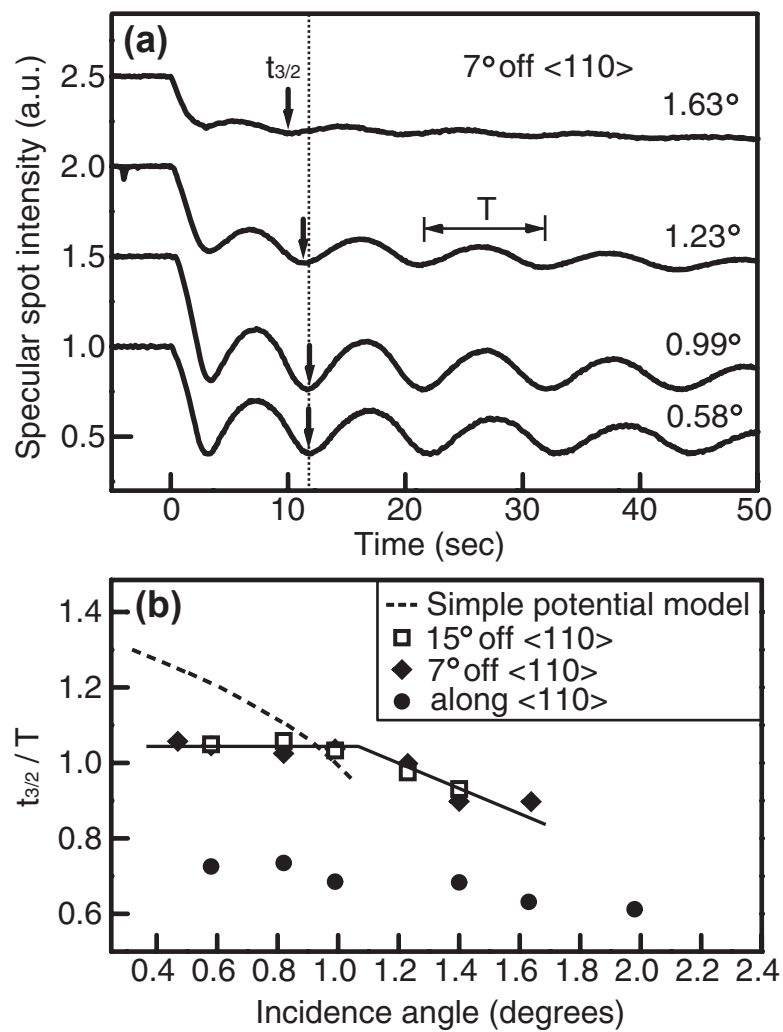


Fig.2

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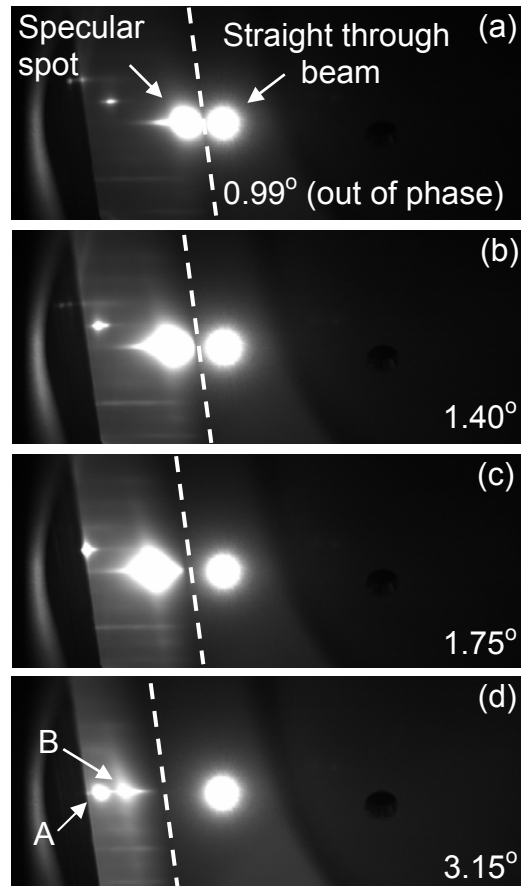


Fig.3

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